

Spin precession and alternating spin polarization in spin-3/2 hole systems

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The spin density matrix for spin-3/2 hole systems can be decomposed into a sequence of multipoles which has important higher-order contributions beyond the ones known for electron systems [R. Winkler, Phys. Rev. B **70**, 125301 (2004)]. We show here that the hole spin polarization and the higher-order multipoles can precess due to the spin-orbit coupling in the valence band, yet in the absence of external or effective magnetic fields. Hole spin precession is important in the context of spin relaxation and offers the possibility of new device applications. We discuss this precession in the context of recent experiments and suggest a related experimental setup in which hole spin precession gives rise to an alternating spin polarization.

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Spin electronics is a quickly developing research area that has yielded considerable new physics and the promise of novel applications [1]. Among the main focuses of spin electronics are semiconductor systems, where ferromagnetic semiconductors and spin polarized transport stand out as major areas of interest. In these fields, the importance of holes as compared with electrons is manifold. Firstly, the compound semiconductors exhibiting ferromagnetism are *p*-type materials, in which ferromagnetism has been shown to be mediated by the itinerant holes [2]. These materials have also been used as sources of spin-polarized holes, motivating the search for a more complete understanding of hole spin precession and relaxation. Secondly, the spin-Hall effect was first studied in the context of a hole Hamiltonian [3, 4] and first observed in a two-dimensional hole gas [5]. With these facts in mind, we devote this Letter to an in-depth study of the spin dynamics of hole systems.

The conduction band of common semiconductors like GaAs is described by a spin-1/2 Hamiltonian so that electron spin dynamics are relatively amenable to theoretical investigation. It is well known that for spin-1/2 electrons the spin-orbit interaction can always be written as a wave vector-dependent effective magnetic field [6, 7]. Electron spin precession in this effective field, as well as in an external field, has been discussed extensively and is well understood [8]. The spin dynamics of holes, described by an effective spin $s = 3/2$ [9], have been studied to a lesser extent [10, 11]. In general, the coupling of the hole spin and orbital degrees of freedom cannot be written as an effective magnetic field, so the simple picture of a spin precessing around an effective Zeeman field is not applicable to holes. Nevertheless, we will show that the spin dynamics of hole systems *can* be viewed as a precession, if precession is understood as a nontrivial *periodic* motion in spin space described by an equation of the type $\frac{d\mathcal{S}}{dt} = \frac{i}{\hbar}[\mathcal{H}, \mathcal{S}]$ for a suitably generalized spin operator \mathcal{S} and spin Hamiltonian \mathcal{H} . While it has long been known

that holes lose their spin information much faster than electrons, to our knowledge a quantitative discussion of hole spin precession based on a firm theoretical footing has not been attempted in the literature. In particular, we will show that in appropriate geometries the spin polarization can alternate in time, even at magnetic field $B = 0$. This alternating spin polarization is a dynamical effect specific to spin-3/2 systems.

The explicit form of the spin-orbit interaction depends sensitively upon the symmetry of the system [12, 13, 14], a fact which is captured in an invariant decomposition of the spin density matrix [15]. Neglecting small terms with cubic symmetry, the spin density matrix of spin-3/2 systems can be expressed in terms of invariants of SU(2). These are proportional to spherical tensors – a monopole, dipole, quadrupole and octupole, denoted respectively by M^0 (a multiple of the identity matrix), \mathbf{M}^1 (the vector of spin-3/2 matrices), \mathbf{M}^2 and \mathbf{M}^3 :

$$\rho = \rho_0 M^0 + \mathcal{S} \cdot \mathbf{M}^1 + \mathcal{Q} \cdot \mathbf{M}^2 + \mathcal{O} \cdot \mathbf{M}^3. \quad (1)$$

The dot product for spherical tensors appearing in Eq. (1) is defined in Ref. [16]. The moments ρ_0 , \mathcal{S} , \mathcal{Q} , and \mathcal{O} provide a set of *independent* parameters characterizing the weights of the multipoles in the expansion (1) [15]. Excepting normalization factors, the monopole moment ρ_0 is the carrier density, while the dipole moment \mathcal{S} corresponds to the spin polarization (Bloch vector) at $B > 0$. The quadrupole moment \mathcal{Q} reflects the splitting between the heavy holes (HHs, spin- z projection $m_s = \pm 3/2$) and the light holes (LHs, $m_s = \pm 1/2$). The octupole moment \mathcal{O} is a unique feature of $s = 3/2$ systems at $B > 0$. We note that in the density matrix of spin-1/2 electrons only the first two terms of the expansion (1) are present. We would like to point out that the term “multipole” is used here in a rather different sense from that of Ref. [4].

The dynamics of spin-3/2 hole systems are determined by the 4×4 Luttinger Hamiltonian \mathcal{H} [9, 17], which, in

the spherical approximation, reads

$$\begin{aligned}\mathcal{H} &= -\frac{\hbar^2\gamma_1}{m_0}\mathcal{K}^0 M^0 - 2\sqrt{5}\mu_B\kappa\mathcal{K}^1 \cdot \mathbf{M}^1 + \sqrt{6}\frac{\hbar^2\bar{\gamma}}{m_0}\mathcal{K}^2 \cdot \mathbf{M}^2 \\ &\equiv \mathcal{H}^0 + \mathcal{H}^1 + \mathcal{H}^2,\end{aligned}\quad (2)$$

where γ_1 , $\bar{\gamma}$, and κ are Luttinger parameters, m_0 is the bare electron mass, and \mathcal{K}^j are tensor operators [15]. In the lowest order of \mathbf{k} and \mathbf{B} we have $\mathcal{K}^0 = k^2$ and $\mathcal{K}^1 = \mathbf{B}$. The operator \mathcal{K}^2 is responsible for the HH-LH coupling in hole systems. We neglect the terms with cubic symmetry, which are small corrections in the present context, as discussed in Refs. [15, 17].

We will analyze in detail the Heisenberg equations of motion (HEM) of the multipoles \mathbf{M}^j . Equation (2) suggests that we study first the HEM

$$\frac{d\mathbf{M}^j}{dt} = \frac{1}{i\hbar} [\mathbf{M}^j, \mathcal{H}^{j'}]. \quad (3)$$

The HEM of a dipole \mathbf{M}^1 evolving under the action of the Zeeman term \mathcal{H}^1 describe the well-known Larmor precession of holes in a magnetic field [18]. These equations are closed, i.e., the RHS does not depend on the other moments $\mathbf{M}^{j''}$, $j'' \neq 1$. In general, however, the HEM for the \mathbf{M}^j in spin-3/2 hole systems cannot be decoupled, having important consequences for hole spin precession. This can be seen from Table I which gives the invariant decomposition of the RHS of Eq. (3) for different j and j' [12]. The most remarkable entry in the table is the one for the quadrupole \mathbf{M}^2 propagating in time due to a quadrupole term in the Hamiltonian, \mathcal{H}^2 , which represents the spin-orbit interaction that gives rise to the HH-LH coupling. The HEM for \mathbf{M}^2 are *not* closed. A quadrupole \mathbf{M}^2 precessing in a quadrupole field “decays” into a dipole \mathbf{M}^1 and an octupole \mathbf{M}^3 . This implies that spin precession of an initially unpolarized system can give rise to spin polarization, even though there is no external or effective magnetic field. [As mentioned above, we use the term spin precession for any HEM (3) with $j, j' > 0$.] Moreover, spin precession of hole systems does not preserve the magnitude of the Bloch vector, as will be shown below explicitly. These observations point to the fact that the spin dynamics of spin-3/2 hole systems are qualitatively different from those of spin-1/2 electron systems, where the HEM for the monopole (number density) is decoupled from that of the dipole (Bloch vector $\boldsymbol{\sigma}$). As a result, electron spin precession preserves the length of the Bloch vector $\langle \boldsymbol{\sigma} \rangle$, i.e., $d|\langle \boldsymbol{\sigma} \rangle|/dt = 0$.

We want to determine and interpret the explicit time evolution of ρ in order to investigate the physics absent in electron systems. This calculation is most easily carried out in the Schrödinger picture, which reflects the equivalence of the Heisenberg and Schrödinger pictures for this problem. In the absence of external fields and disorder, the density matrix satisfies the quantum Liouville equation $\frac{\partial \rho}{\partial t} = \frac{i}{\hbar} [\rho, \mathcal{H}]$, with formal solution

TABLE I: Irreducible representations $\mathcal{D}^{J''}$ of $SU(2)$ of the (linear combinations of) multipoles $\mathbf{M}^{J''}$ contained in an invariant decomposition of $(1/i\hbar) [\mathbf{M}^j, \mathcal{H}^{j'}]$.

	\mathcal{H}^0	\mathcal{H}^1	\mathcal{H}^2	\mathcal{H}^3
\mathbf{M}^0	0	0	0	0
\mathbf{M}^1	0	\mathcal{D}^1	\mathcal{D}^2	\mathcal{D}^3
\mathbf{M}^2	0	\mathcal{D}^2	$\mathcal{D}^1 + \mathcal{D}^3$	\mathcal{D}^2
\mathbf{M}^3	0	\mathcal{D}^3	\mathcal{D}^2	$\mathcal{D}^1 + \mathcal{D}^3$

$\rho(t) = e^{-i\mathcal{H}t/\hbar}\rho(0)e^{i\mathcal{H}t/\hbar}$, where $e^{i\mathcal{H}t/\hbar}$ is the time evolution operator (which can often be evaluated in closed form). We consider first an example where we assume the hole spin to be oriented initially along the z -direction, $m_s = +3/2$, so that

$$\tilde{\rho}(t=0) = \frac{1}{2}M_0^0 + \frac{3}{2\sqrt{5}}M_0^1 + \frac{1}{2}M_0^2 + \frac{1}{2\sqrt{5}}M_0^3, \quad (4)$$

where the tilde indicates that ρ has been normalized with respect to the total density $2\rho_0$. This equation demonstrates that, in general, the density matrix of holes cannot be written simply as the sum of a monopole and a dipole. The higher multipoles will be present [19]. We want to restrict ourselves to $\mathcal{H} = \mathcal{H}^0 + \mathcal{H}^2$, i.e., $B = 0$. Table I shows that ρ evolves as a combination of a dipole, a quadrupole, and an octupole. The implications of this fact for the spin are seen by following the motion of the Bloch vector $\mathbf{S}(t) = \frac{3}{2\sqrt{5}}\tilde{\mathbf{S}}(t)$

$$\begin{aligned}\mathbf{S}(t) &= \frac{3}{2}\left\{ \hat{\mathbf{S}}_0 [\cos^2(\omega t) + c^4 \sin^2(\omega t)] \right. \\ &\quad + \hat{\mathbf{k}} c s (1 + c^2) \sin^2(\omega t) \\ &\quad \left. + (\hat{\mathbf{S}}_0 \times \hat{\mathbf{k}}) 2 c s \sin(\omega t) \cos(\omega t) \right\},\end{aligned}\quad (5)$$

where the unit vector $\hat{\mathbf{S}}_0$ is the orientation of \mathbf{S} at $t = 0$; $\omega = \bar{\gamma}\hbar k^2/m_0 = (\varepsilon_h - \varepsilon_l)/2\hbar$, with ε_h and ε_l the HH and LH energies, respectively; $c = \hat{\mathbf{S}}_0 \cdot \hat{\mathbf{k}}$ is the cosine of the angle between $\hat{\mathbf{S}}_0$ and $\hat{\mathbf{k}}$; and s is the sine of the same angle. When \mathbf{S}_0 is parallel to \mathbf{k} (i.e., $c = 1$), we get $\mathbf{S}(t) = \mathbf{S}_0$, which is due to the fact that the initial state is an eigenstate of the Hamiltonian. In general, neither the magnitude nor the orientation of the Bloch vector are conserved. This is illustrated in Fig. 1 showing $\mathbf{S}(t)$ for an angle of 60° between $\hat{\mathbf{S}}_0$ and $\hat{\mathbf{k}}$. Helicity $\mathbf{S} \cdot \mathbf{k}$ is conserved, a well-known fact about this model, which sheds additional light on spin precession in hole systems. Since $\frac{d}{dt}(\mathbf{S} \cdot \mathbf{k}) = 0$ and the wave vector is not changing, $\frac{d\mathbf{S}}{dt} \cdot \mathbf{k} = 0$. Therefore, whenever the magnitude of the spin changes, the angle between spin and wave vector must change in order to preserve the projection of \mathbf{S} onto \mathbf{k} . As a consequence, no nontrivial spin precession occurs when $\mathbf{S} \perp \mathbf{k}$ (or $\mathbf{S} \parallel \mathbf{k}$). We note that energy is also conserved for spin precession in hole systems. Yet for $B > 0$, when $\mathcal{H} = \mathcal{H}^0 + \mathcal{H}^1 + \mathcal{H}^2$, it can be shown that,

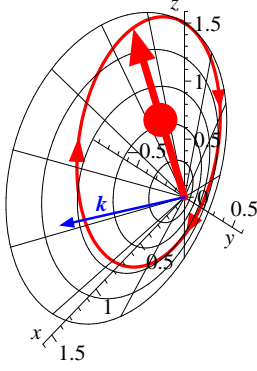


FIG. 1: Precession of the Bloch vector \mathbf{S} (bold arrow) about the wave vector \mathbf{k} when the angle between the two is initially 60° . We have assumed $\mathbf{S}(t=0)$ is parallel to the z axis, and \mathbf{k} lies in the xz plane.

in the general case, energy is transferred back and forth between \mathcal{H}^1 and \mathcal{H}^2 as time progresses.

We would like to relate our work to recent experiments by Grayson *et al.* [20, 21] which demonstrated that a high-quality bent heterostructure can be grown on top of a pre-cleaved corner substrate that allows one to drive the charge carriers around an atomically sharp 90° corner. Grayson's experiments were performed on a two-dimensional (2D) electron system in a GaAs/AlGaAs quantum well. We show here that a similar system containing holes gives rise to fascinating new physics. The setup is sketched in Fig. 2(a). We assume $B = 0$ so that the Hamiltonian is $\mathcal{H} = \mathcal{H}^0 + \mathcal{H}^2$. An unpolarized HH wave packet travels in the 2D channel L_1 in the $+x$ direction. For simplicity we assume that all orbital degrees of freedom are integrated over, so that ρ depends only on the spin indices and does not depend on the wave vector \mathbf{k} . Nevertheless, the coupling of spin and \mathbf{k} is present in the Hamiltonian through \mathcal{H}^2 . The magnitudes of the normalized moments at $t \leq 0$ (i.e., before reaching the corner) are $\tilde{S} = \tilde{O} = 0$ and $\tilde{Q} = 1/2$. The spin quantization axis of the HH states in L_1 is parallel to the z -direction. After the wave packet has passed the corner, the HH states are not eigenstates of \mathcal{H}^2 , their spin quantization axis being perpendicular to the spin quantization axis supported by the quadrupole field in L_2 . Therefore, the quadrupole and octupole moments in L_2 oscillate in time

$$|\tilde{Q}(t)|^2 = (1/16) + (3/16) \cos^2(\omega_z t) \quad (6a)$$

$$|\tilde{O}(t)|^2 = (3/16) \sin^2(\omega_z t), \quad (6b)$$

with precession frequency $\omega_z = (\varepsilon_h - \varepsilon_l)/2\hbar \simeq 2\tilde{\gamma}\hbar\pi^2/m_0w^2$. This frequency can be tailored by varying the width w of the 2D channel. For GaAs in the spherical approximation we have $\tilde{\gamma} = 2.58$ and we take $w = 10$ nm, yielding a precession period $2\pi/\omega_z \approx 11$ ps.

For the simplified geometry of Fig. 2(a) the spin polarization \tilde{S} in L_2 remains zero, as required by the con-

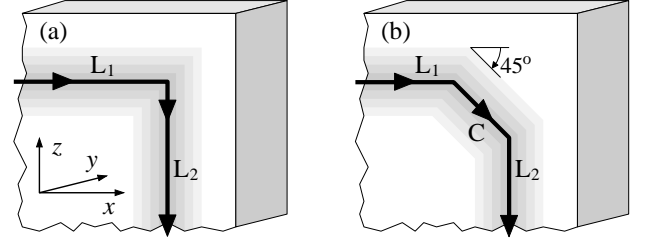


FIG. 2: A bent structure allows holes to be driven around a corner [20, 21]. (a) An idealized representation of the corner device. (b) A more realistic model of the corner, as discussed in the text. In regions C and L_2 the spin polarization oscillates as a function of time.

servation of helicity discussed above. However, assuming a sharp 90° corner for an *ideal* 2D system is certainly an oversimplification, even when the corresponding *quasi* 2D system has atomically sharp interfaces [20]. A more realistic treatment can be obtained by modeling the transition region between the channels L_1 and L_2 as a sequence of two 45° corners as sketched in Fig. 2(b). Once again, an unpolarized HH wave packet travels in channel L_1 in the $+x$ direction, the initial conditions being the same as in the previous example. If the HH wave packet enters the central region C at $t = 0$, we get for the squared normalized moments in this region

$$|\tilde{S}(t)|^2 = (9/80) \sin^2(\omega_z t) \quad (7a)$$

$$|\tilde{Q}(t)|^2 = 1/64 + (15/64) \cos^2(\omega_z t) \quad (7b)$$

$$|\tilde{O}(t)|^2 = (39/320) \sin^2(\omega_z t). \quad (7c)$$

Equation (7a) shows that the initially unpolarized hole current acquires an alternating spin polarization $\tilde{S}(t)$ due to spin precession at $B = 0$. In Cartesian coordinates, the Bloch vector in region C reads $\tilde{S}(t) = [0, -\frac{3}{4\sqrt{5}} \sin(\omega_z t), 0]$. When the HH wave packet enters the channel L_2 it continues to precess. We get for the spin polarization

$$\tilde{S}_y = \frac{-3}{8\sqrt{5}} \left[\cos^2 \left(\frac{\omega_z T}{2} \right) \sin(\omega_z t) + 2 \sin(\omega_z T) \sin^2 \left(\frac{\omega_z t}{2} \right) \right]. \quad (8)$$

Here T is the time required to traverse C which depends on the length of C and the magnitude of the in-plane wave vector. It determines the fraction of the spin polarization in L_2 that will be oscillating. If we take the length of C to be of the order of the channel width, namely $w = 10$ nm, and the initial wave vector $k_F = 0.1$ nm $^{-1}$, we get $T \sim 0.5$ ps. The amplitude of \tilde{S}_y in this case is approximately 0.1. We omit here the qualitatively similar but more complicated expressions for $\tilde{Q}(t)$ and $\tilde{O}(t)$. We note also that the approach in Fig. 2(b) can be further extended in a transfer-matrix-like approach in order to describe more complicated geometries.

We expect hole spin precession to be robust against disorder provided $\omega_z > 1/\tau_s$, a range experiment suggests

is realistic. Hole spin relaxation times τ_s in the range of tens of ps [22] or even one ns [23] have been observed experimentally for GaAs quantum wells, demonstrating that the spin precession discussed in this paper cannot be neglected in the context of hole spin dynamics. We expect that the precession can be observed experimentally using time-resolved optical techniques such as Kerr or Faraday rotation spectroscopy.

The examples above identify an interesting potential application of spin-orbit effects in hole systems. By choosing appropriate initial conditions the spin polarization in the bulk of the system can be made to oscillate as a function of time. This oscillating spin polarization is to be contrasted with the Zitterbewegung discussed lately [24], in which the carrier *position* oscillates as a function of time. Unlike conventional transistors, which work in the diffusive limit, the examples we discuss are valid in the ballistic limit, a feature shared with the Datta spin transistor [25]. The situation we describe is nevertheless different from the spin precession in a Datta spin transistor where the electrons precess not as a function of time but as a function of position [8, 25].

The above example illustrates the *controlled* hole spin precession in a confined geometry in a ballistic regime, where the holes experience an essentially unidirectional quadrupole field. The situation is qualitatively different in, e.g., bulk material in a diffusive regime, where the occupied hole states are characterized by many different orientations of the wave vector \mathbf{k} . The quadrupole field is thus randomly oriented so that spin precession in this field must also be taken into account for spin relaxation, similar to Dyakonov-Perel (DP) spin relaxation in electron systems. Previously [26, 27], spin relaxation of hole systems was discussed mostly using an Elliot-Yafet-type approach [7], where spin relaxation is caused by momentum scattering events. DP spin relaxation, on the other hand, implies that the spin orientation is lost due to spin precession *in between* the momentum scattering events. Such spin precession for the case of holes has been described by Averkiev *et al.* [28] but only as a result of the spin splitting induced by bulk and structure inversion asymmetry in 2D systems. On the other hand, the spin precession described by Eq. (5) is different in nature, being due to the quadrupole field \mathcal{H}^2 so that it is present also in inversion symmetric systems. Note also that hole spin precession due to \mathcal{H}^2 is typically one or several orders of magnitude faster than the precession in the effective magnetic field due to a broken inversion symmetry so that \mathcal{H}^2 can yield the more important contribution to DP hole spin relaxation. (We use here the term DP spin relaxation in a generalized sense for any relaxation based on spin precession, even if motional narrowing [7] is not important.) Equation (5) suggests that hole DP spin relaxation in bulk systems occurs on time scales of the order of $2\pi\omega^{-1} = 2\pi(2\gamma\hbar k^2/m_0)^{-1}$. In bulk GaAs, for holes optically excited with light at ~ 800 nm, recent

experiments yielded spin relaxation times of about 110 fs [29], while the simple estimate above gives ~ 200 fs for this case. (This number depends sensitively on the wave vector of the occupied hole states that can only be estimated for the experiments in Ref. [29].) We remark that, in analogy with spin relaxation in anisotropic electron systems [28, 30], the spin relaxation of hole systems will be characterized by different relaxation times for each multipole moment in the density matrix.

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